Superconducting Transition of ErBaCuO+x

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Superconducting Transition of $ErBa_2Cu_3O_{6+x}$

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The magnetic response of $ErBa_2Cu_3O_{6+x}$ has been studied in terms of complex susceptibility $\chi = \chi' - i\chi''$. In the superconducting transition, χ' reflects the Meissner effect and χ'' forms a single peak. Careful measurements of a disk sample revealed that the onset temperature of χ' is slightly higher than that of χ'' and there exists two superconducting phases. χ' and χ'' of the lower phase are very sensitive to the field amplitude. The higher phase may be interpreted as a bulk superconductivity while the lower phase corresponds to a weak coupling between bulk superconducting inclusions. Above discussion is confirmed by measurements of a pulverized sample.

KEY WORDS: Oxide superconductor/ Complex susceptibility/ Transition temperature/ Hartshorn bridge/

I. INTRODUCTION

Since the discovery of high-T_c superconductors, many people in the field have been making a considerable effort to unveil their superconductive behavior. Generally, the resistance zero does not ensure the bulk nature of superconductivity. For this purpose, the measurement of magnetic response becomes important. The dc susceptibility measurement led to the conclusion that this superconductivity has a bulk nature. However, it should be pointed out that the perfect diamagnetism has not been observed for high-T_c oxide superconductors with a dc SQUID susceptometer. The complex susceptibility offers an alternative way of examining magnetic characteristics of a superconductor.

In our previous works, we studied the magnetic response of $La_{1.9}Sr_{0.1}CuO_{4-x}$,[1,2] $YBa_2Cu_3O_{7-x}$,[3] and $GdBa_2Cu_3O_{7-x}$.[4] By means of the complex susceptibility, it has been revealed that these oxide superconductors prepared by the sintering treatment exhibit two phases in the superconducting transition, i.e., a fairly solid superconducting phase and a weakly-coupled superconducting phase.

In this work, we report detailed study in terms of complex susceptibility on another 90–K class oxide superconductor, $ErBa_2Cu_3O_{6+x}$.

II. EXPERIMENTAL

A. Sample Preparation

The $ErBa_2Cu_3O_{6+x}$ samples were prepared from the solid state reaction starting from the mixture of Er_2O_3, $BaCO_3$, and CuO. The mixture was reacted at 850°C for 5 h in air. The pelletized sample was subsequently sintered at 850°C for 17 h, at 900°C for 5 h, and at 950°C

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for 6.5 h. The sample was cooled slowly in the furnace. The specimen diameter decreased by 12% by the 950°C sintering. Sintering at 1000°C caused a partial melting of the specimen and by the 900°C sintering the sample did not shrink appreciably but easily absorbed a drop of alcohol. From the X-ray powder diffraction pattern, our specimen was found to be single phase orthorhombic.[5] Compared to the density estimated from the lattice constant, the real density reached 83%. Oxygen contents of the sample were not determined.

In order to extract a bulk phase in a more direct method, we prepared a powdered specimen dispersed in an insulating matrix. One of the sintered pellets was crused and pulverized thoroughly. The Al_2O_3 powder, as an insulator, was also ground in an agate mortar. The 1:2 mixture (in weight) of $ErBa_2Cu_3O_{6+x}$ and Al_2O_3 powder was again ground in the mortar and pressed into a pellet.

B. Measurements of χ' and χ''

The measuring system of the superconducting transition by means of complex susceptibility, $\chi' - i\chi''$, consists of the Hartshorn bridge and a temperature control system (1–325 K). The details of our device were previously reported.[2,6,7] Since the bridge-balance condition depends on the coil temperature, we made the 4.2–100 K measurement at a coil temperature of 4.2 K (immersed in a liquid He bath) and the 80–100 K measurement at a coil temperature of 77 K (immersed in a liquid N_2 bath).

A null adjustment of the bridge was made at the sample temperature of 105 K. Phase setting of the lock-in analyzer was made by the so-called off-balance method.[1] Temperature was measured with a calibrated carbon-glass thermometer. For the measurement of a disk sample, the ac magnetic field $h(t) = h_0 \sin 2\pi ft$ was applied perpendicular and parallel to the flat surface of the disk. For the measurement of a powdered sample, the direction of the applied field should not be essential.

III. RESULTS AND DISCUSSION

Typical transition curves of χ' and χ'' are shown in Fig. 1, where $h_0 = 500$ mOe and $f = 132$ Hz. In Figs. 2 and 3, we present $-\chi'$ and χ'' as a function of T for several different h_0 (5–1500 mOe), where f is fixed at 132 Hz. The characteristic features of our findings are itemized as follows.

(a) The smooth transition of χ' corresponds to the Meissner effect. The transition is very sharp at 5 mOe (0.7 K for 10–90% change). Careful examinations of χ' curve make one notice two (higher and lower) superconducting phases.

(b) χ'' forms a single peak in the transition region. However, the onset temperature of χ'' is lower than that of χ' by 0.5 K (typically), supporting the existence of two phases.

(c) As h_0 increases, the higher-temperature portion of the χ' transition curve converges onto a single line.

(d) As h_0 increases, the peak height of χ'' increases and then decreases. Note that the h_0-dependent growth of the χ'' peak was observed for some inhomogeneous superconductors[7] and lower-dimensional superconductors,[8] but the growth is always monotonic with h_0.

(e) There was no frequency dependence of χ' and χ'' between 50 and 320 Hz.

These experimental findings may be a clue to explain the complex-susceptibility behavior

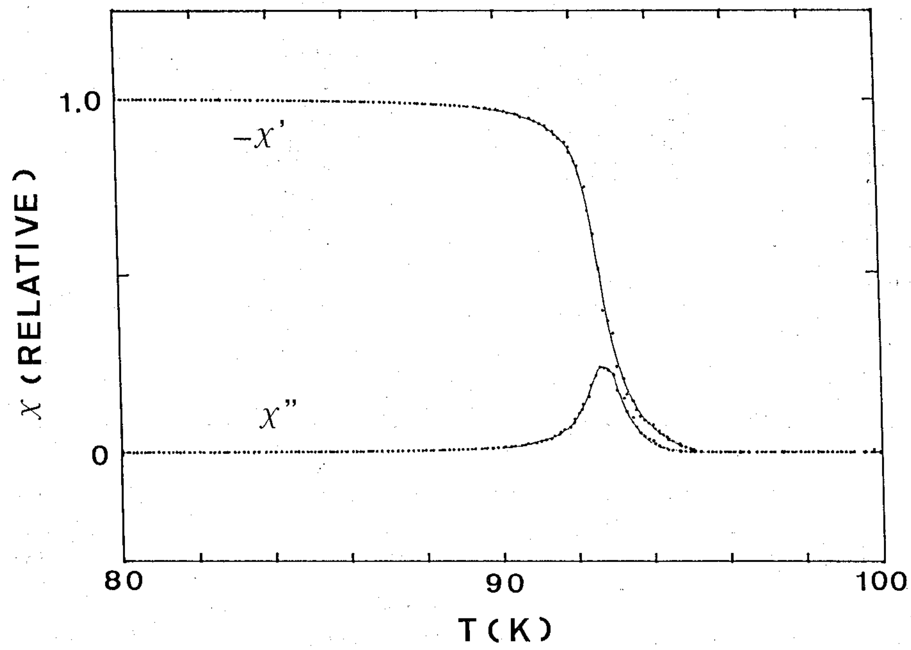

Fig. 1. Real and imaginary components of the susceptibility. The ac field is perpendicular to the flat surface of the sample, where $h_0=500$ mOe and $f=132$ Hz.

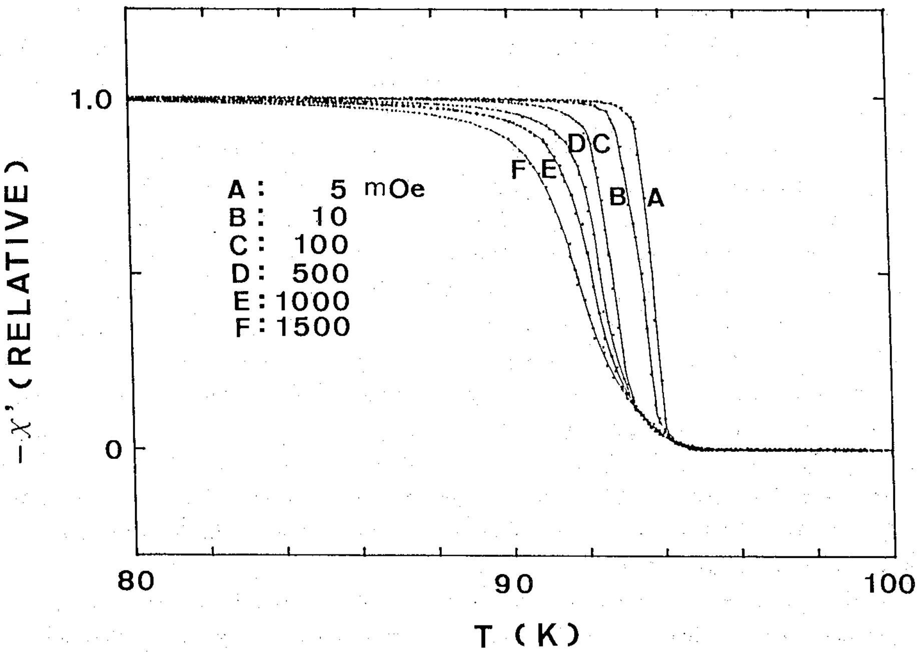

Fig. 2. h_0 dependence of the real component of the susceptibility, where $f=132$ Hz.

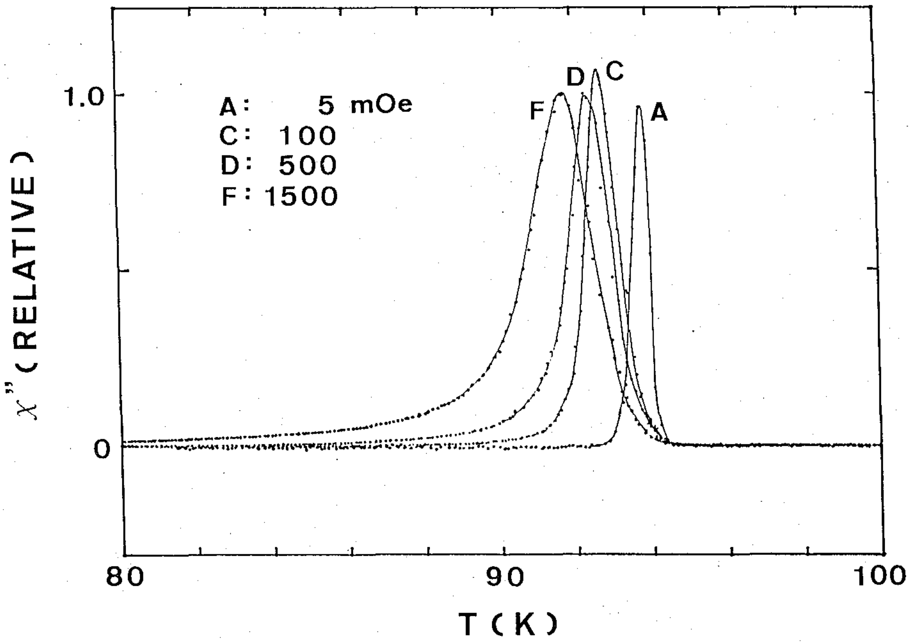

Fig. 3. h_0 dependence of the imaginary component of the susceptibility, where f=132 Hz. Some data are omitted for clarity.

of $ErBa_2Cu_3O_{6+x}$. First the insensitivity of χ to f rules out the possibility of employing an effective conductivity model[9] to understand these features. Second, if we explain these only by a weakly-connected loop model[7] or something similar, there is a serious difficulty at onset temperatures. According to the model, $-d\chi''/d\chi'$ diverges at the onset temperature (see Eq. 3 of Ref. 7). This means that χ'' has to be larger than $-\chi'$ at temperatures near the onset. The experimental fact is the opposite (see Fig. 1). Third, the above item (c) suggests the existence of a solid superconducting phase. Being responsible for a bulk superconductivity, the dc susceptibility may be sensitive to this phase. Thus we infer this phase does not increase to fulfill the perfect diamagnetism as T goes down. Fourth, as mentioned in our previous work,[1] the weak couplings may occur in the lower phase. Therefore, the h_0-sensitive profile of χ' and the peak formation of χ'' are plausible from the spirit of the weakly-connected loop model.

To confirm the above discussion, we extend the measurement to the powdered $ErBa_2Cu_3O_{6+x}$ specimen. Figure 4 presents χ', χ'' versus T of the powdered specimen for 10 mOe and 1000 mOe. The χ' curve is very similar to the prediction for a bulk-phase contribution to the complex susceptibility. As T decreases further from 80 K, $-\chi'$ increases monotonically and reaches 1.26 times of 80-K value at 30 K (not shown in Fig. 4). One notices a slight descrepancy of χ' curves between 10 and 1000 mOe below 90 K. The peak in the χ'' curve completely disappears, meaning that the coupled nature in the specimen is largely suppressed by the pulverization.

In Fig. 5 we give the comparative presentation of χ' versus T for perpendicular, parallel, and powdered cases, where the vertical scale is normalized to the weight of $ErBa_2Cu_3O_{6+x}$. One notices a coincidence of the χ' curves at temperatures near the onset, where the demagnetization effect is not expected to differ remarkably in the three cases. At lower

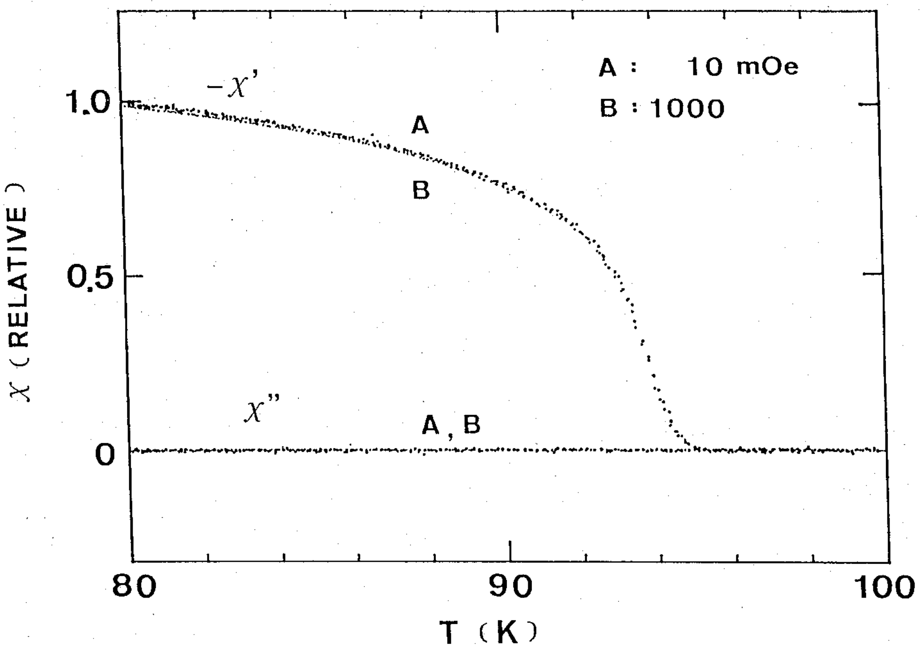

Fig. 4. Complex susceptibility vs temperature for the powdered sample. The dependence of the real component on h_0 is quite small. No imaginary component is visible.

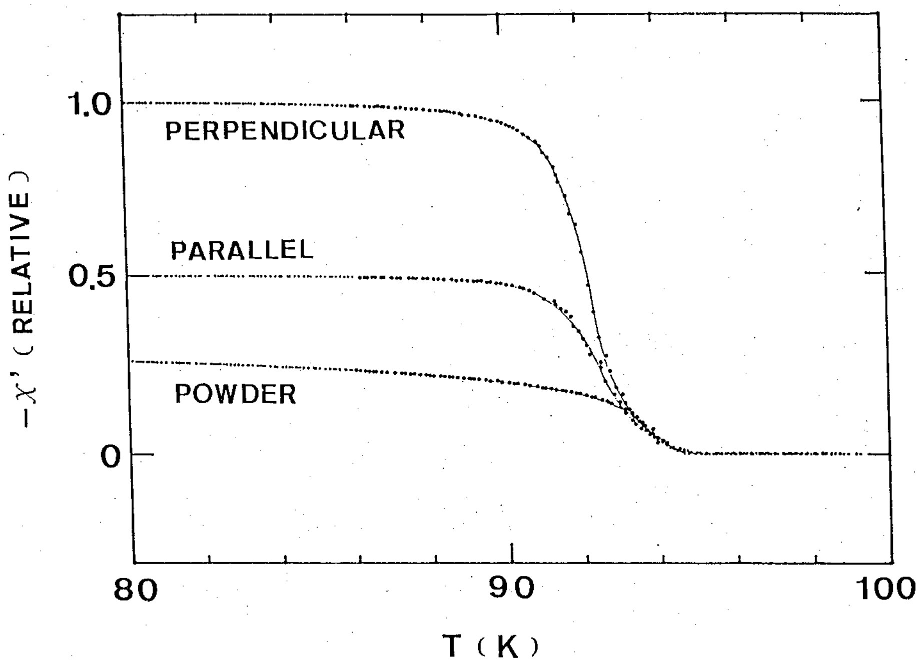

Fig. 5. Real component of the susceptibility with temperature for the sintered pellet and the powdered sample, where $h_0=1000$ mOe and $f=132$ Hz. Data are normalized by the weight of $ErBa_2Cu_3O_{6+x}$ in each sample.

temperatures, the saturated values of χ' are considerably different from each other. We consider that only the bulk phase is observed in the powdered case. It is not unreasonable to assume that the powdered specimen consists of spheres, of which the demagnetization factor is 1/3. For the sintered spherical specimen, the χ' value to be compared with that of the powdered specimen should be located in between the perpendicular and parallel cases at 80 K. Under this assumption, we estimate that the 26–52% volume fraction of the sintered pellet shows a bulk superconductivity at 80 K. This is in accordance with the result of the dc susceptibility.[10)]

The present results as well as our previous findings with other oxide superconductors,[1-4)] can be summerized as the $\chi'-T$ relation (see Fig. 6), where the bulk- and coupled-phase contributions are separately drawn. T_1 indicates the onset temperature of χ' and T_2 is the onset of coupled phase. Experimentally, T_2 corresponds to the onset temperature of χ''. As h_0 increases, only the coupled phase would be influenced and the situation of item (c) is well illustrated in this figures. It is plausible to consider that the lower-phase component $\Delta\chi_L'$ is responsible for the peak formation of χ''. At larger h_0 the transition region shifts toward lower temperatures, resulting in a decrease of $\Delta\chi_L'$. This is the reason why the χ'' peak does not grow monotonically with h_0.

The measurement of the powdered sample at lower temperatures exhibits an interesting magnetic response. As the temperature is lowered, $-\chi'$ increases monotonically down to about 10 K. However, near that temperature, $-\chi'$ takes the maximum value and then decreases slightly with the decrease of temperature (see Fig. 7). This turning of the $-\chi'$ curve is probably due to the field-induced paramagnetism which is expected to be enhanced at very low temperatures. As pointed out by Thompson et al.,[11)] the result suggests that field-induced paramagnetism and superconductivity exist independent of one another.

Confirmation of the induced paramagnetism of Er^{3+} ions was made with a tetra-$ErBa_2Cu_3O_{6+x}$ powdered specimen (prepared by quenching from 950°C to 77 K, not supercon-

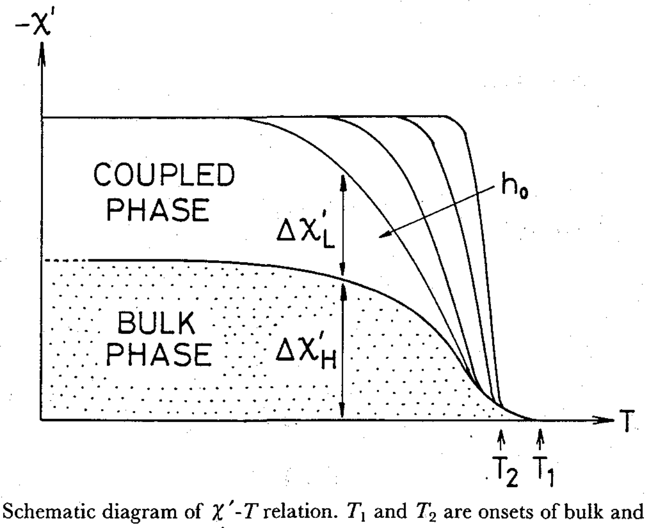

Fig. 6. Schematic diagram of χ'-T relation. T_1 and T_2 are onsets of bulk and coupled phases, respectively. χ' is divided into coupled- and bulk-phase components $\Delta\chi_L'$ and $\Delta\chi_H'$. h_0 increases along the arrow.

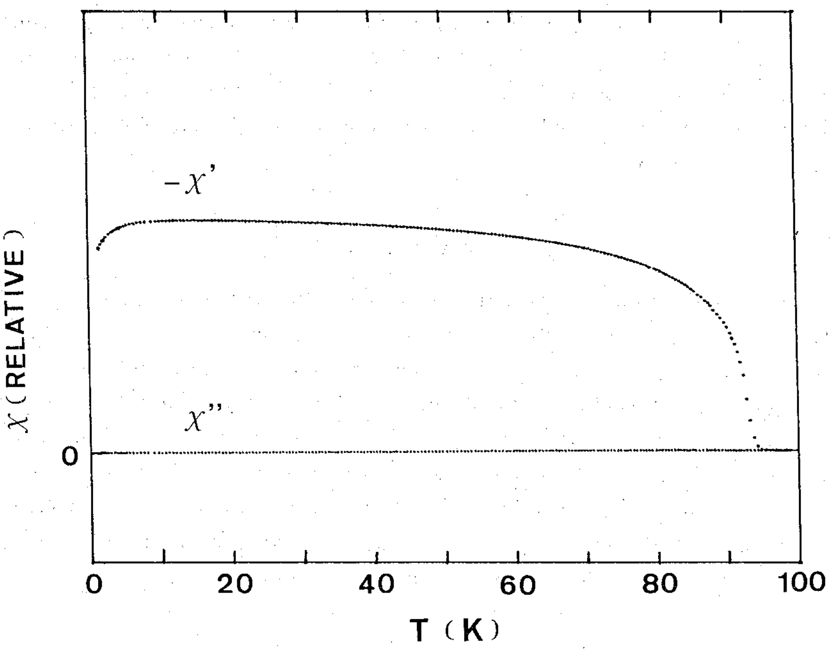

Fig. 7. Real and imaginary components of the susceptibility vs temperature for a powdered sample. The diameter of the particles is less than 25 μm.

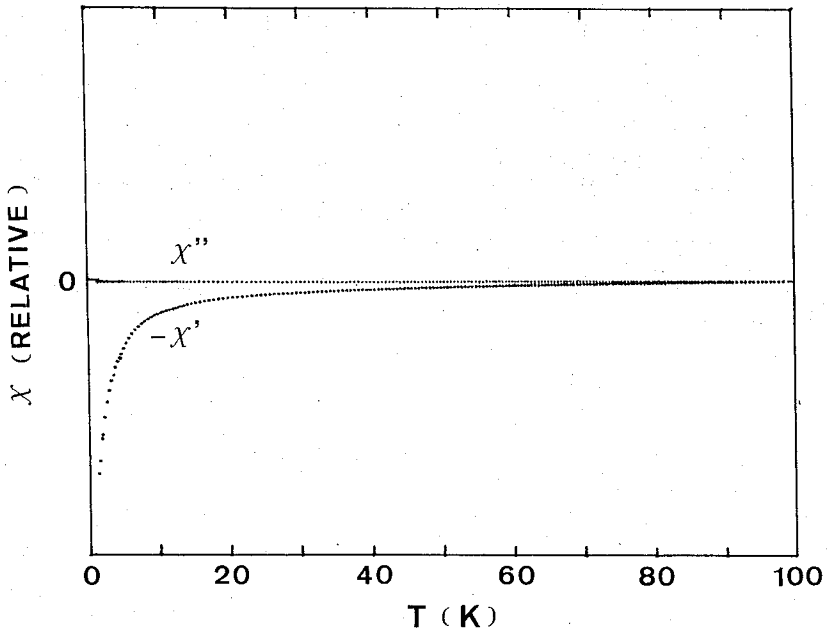

Fig. 8. Real and imaginary components of the susceptibility vs temperature for a tetra-phase powdered sample. The rapid decrease of $-\chi'$ curve at very low temperatures is due to the paramagnetic effect of Er^{3+} ions.

ducting). As shown in Fig. 8, $-\chi'$ begins to decrease smoothly below 80 K and then rapid decrease takes place below about 10 K, certainly due to the enhanced paramagnetism of Er^{3+} ions. It should be noted that the turning point observed for a powdered sample was not visible for a disk sample. The reason for this is that in the disk sample, the disk sample, the field-induced paramagnetism hardly takes place due to the diamagnetic shielding current covering the whole specimen.

VI. CONCLUSIONS

We measured the complex susceptibility of the $ErBa_2Cu_3O_{6+x}$ samples. The plausible conclusion is that the superconducting transition of a disk sample is built by the subtle balance between bulk and coupled phases. The h_0-dependent profile of the complex susceptibility is useful in examining the bulk nature of these samples. Measurement of the pulverized specimen also supports this idea. The coupled phase can be seen as a peak formation inb χ''. It is well known that the eddy-current loss possibly gives a peak formation in χ'' as well. In this regard, the frequency dependence of χ is also essential for examining the superconductive nature. From measurment of the pulverized sample, we also confirmed that field-induced paramagnetism and superconductivity exist independent of one another.

REFERENCES

(1) H. Mazaki, M. Takano, R. Kanno, and Y. Takeda, *Jpn. J. Appl. Phys.* **26**, L780 (1987).
(2) H. Mazaki, M. Takano, Z. Hiroi, Y. Bando, R. Kanno, Y. Takeda, and O. Yamamoto, *Bull. Inst. Chem. Res., Kyoto Univ.*, **65**, 147 (1987).
(3) H. Mazaki, M. Takano, Y. Ikeda, Y. Bando, R. Kanno, Y. Takeda, and O. Yamamoto, *Jpn. J. Appl. Phys.*, **26**, L1749 (1987).
(4) H. Mazaki, M. Takano, R. Kanno, and Y. Takeda, *Jpn. J. Appl. Phys.*, **26**, L1752 (1987).
(5) T. Ishida, *Jpn. J. Appl. Phys.*, **26**, L1294 (1987).
(6) T. Ishida and H. Mazaki, *Phys. Rev. B*, **20**, 131 (1979).
(7) T. Ishida and H. Mazaki, *J. Appl. Phys.*, **52**, 6798 (1981).
(8) T. Ishida, K. Kanoda, H. Mazaki, and I. Nakada, *Phys. Rev. B*, **29**, 1183 (1983).
(9) E. Maxwell and M. Strongin, *Phys. Rev. Lett.*, **10**, 212 (1963).
(10) H. Takagi, S. Uchida, H. Sato, H. Ishii, K. Kishio, K. Kitazawa, K. Fueki, and S. Tanaka, *Jpn. J. Appl. Phys.*, **26**, L601 (1987).
(12) J. R. Thompson, S. T. Sekula, D. K. Christen, B. C. Sales, L. A. Boatner, and Y. C. Kim, *Phys. Rev. B*, **36**, 718 (1987).